Cirrus clouds in the tropical tropopause layer: 
Role of heterogeneous ice nuclei

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Received 20 February 2004; revised 11 May 2004; accepted 19 May 2004; published 16 June 2004.

[1] The role of heterogeneous ice nuclei in controlling the occurrence and properties of cirrus clouds in the tropical tropopause layer is examined with the help of a Lagrangian microphysical cirrus model that includes competition between insoluble and volatile aerosol particles during ice nucleation and small-scale temperature perturbations. The potential of ice nuclei to influence the moisture budget of air at entry to the stratosphere appears to be limited. Additional dehydration of up to 0.3 ppmv may occur, relative to 1.3 ppmv caused by pure homogeneous freeze-drying. In contrast, ice nuclei significantly enhance the frequency of occurrence of subvisible cirrus clouds, even when present at concentrations as low as 0.01 L⁻¹. This is likely to be the largest effect of ice nuclei on cirrus near the tropical tropopause. Ice nuclei may also alter cloud radiative forcing by changing the ice water content, increasing the effective crystal radius, and decreasing the cloud lifetime.


1. Introduction

[2] The future evolution of ozone and climate is linked to processes controlling stratospheric humidity. Dynamical and microphysical processes occurring in the tropical tropopause layer (TTL) set the boundary condition for stratospheric water vapor (H₂O). Details of these processes and their relative roles are not well understood. Optically thin and subvisible cirrus (SVC) clouds form in situ in the TTL and dehydrate the air entering the stratosphere by sedimenting ice crystals. The efficiency of dehydration by SVCs and resulting effects on the distribution of relative humidity over ice (RHI) depends on lifetimes and properties of ice crystals, which in turn are controlled by horizontal and vertical air motions and the mechanisms that initiate the ice phase [Holton and Gettelman, 2001; Jensen et al., 2001; Clark et al., 2003; Luo et al., 2003; Murphy, 2003; Jensen and Pfister, 2004].

[3] There is mounting observational evidence for an impact of heterogeneous ice nuclei (IN) on cirrus clouds followed by homogeneous freezing [Haag et al., 2003; DeMott et al., 2003]. These studies suggest background IN concentrations ≤10–30 L⁻¹ at northern midlatitudes, obtained by inference from measurements of RHI and by direct measurements of IN, respectively. Ice nuclei could possibly play a role in determining cirrus properties in the TTL [Kärcher, 2002]. They could trigger ice formation at values RHIIN lower than required for homogeneous freezing, thereby enhancing the downward flux of H₂O and causing additional dehydration.

[4] Number concentration and freezing RHI (as a surrogate for chemical composition) of IN are the key quantities controlling the IN effect on cirrus. Only limited information from direct measurements about these quantities in TTL conditions is available. The IN number density, nIN, required to impact SVCs may be much smaller than the values found in the lower troposphere near IN source regions (up to ~10⁴ L⁻¹). Deep convection and large-scale equatorward transport could import IN into the TTL despite efficient wet removal. Metallic and mineral dust particles have been found enriched in cirrus ice crystal residues and could serve as IN [DeMott et al., 2003]. Insoluble meteoric material of stratospheric origin has been found in ice crystals in substropical anvil cirrus [Cziczo et al., 2004] and might serve as efficient IN. Carbonaceous aerosols from biomass burning or forest fires and other midlatitude sources might enter the tropical upwelling region, but the efficacy of smoke and soot particles to nucleate ice is currently unclear.

[5] Gettelman et al. [2002] parameterized crystal formation and fallout which does not permit to distinguish between different freezing pathways. Jensen and Pfister [2004] varied the freezing RHI of liquid particles in a detailed model but did not include IN as a separate particle type, hence cirrus formation was not limited by nIN. It is the aim of the present study to explore the possible impact of IN on cirrus, ice supersaturation, and dehydration in the TTL, taking the key unknowns nIN and RHIIN as free parameters. Effects of transport processes on the TTL moisture budget are not considered. Our results help define the range of possible microphysical processes influencing stratospheric humidity.

2. Model Approach

[6] We use the detailed Lagrangian microphysical model APSCM [Kärcher, 2003] to simulate aerosol particle growth and evaporation, ice crystal nucleation, growth, sublimation, and sedimentation along trajectories in the TTL. One version of the model has recently been applied to study the role of IN in midlatitude cirrus [Haag and Kärcher, 2004]. Vertical advection of ice crystals is parameterized by a size-dependent exponential removal of ice...
1–2 km below the thermal tropopause located at ultimately ends up in the stratosphere occur within the TTL clouds and dehydration of possibly supersaturated air that vapor mixing ratio, [H2O], of 4 ppmv. For each simulation, altitude. Our model domain thus lies within 365–380 K of 800 cm a lognormal distribution with a total number concentration [51x121] although particles may contain other species as well mainly consist of aqueous sulfuric acid [51x341] large-scale cooling may be associated with the passage of air from the edge to the center of the cold trap.

[s] Vertical air motions driven by mesoscale gravity waves are known to play a crucial role in the formation of tropical and midlatitude cirrus [Jensen et al., 2001; Kärcher and Ström, 2003]. Neglecting small-scale variability in cooling rates would overestimate the role of IN in cirrus formation. To account for small-scale dynamic features, we superimpose stochastic T-perturbations (+0.5 K amplitudes, <1 h periods) to the slowly cooling trajectories, resulting in a spectrum of adiabatic cooling rates that maximizes near 10 K h^{-1} and is skewed towards lower values. This results in different cooling rates at the time of freezing and thus in dynamically-induced variations of cloud properties, but at the same time allows us to isolate the effects of IN. We are not attempting to reproduce real air parcel histories, but rather perform a statistical analysis.

[9] Aerosol particles in the upper tropical troposphere mainly consist of aqueous sulfuric acid [Brock et al., 1995], although particles may contain other species as well [Murphy et al., 1998]. These aerosols are initialized with a lognormal distribution with a total number concentration of 800 cm^{-3}, a mean dry number radius of 0.075 µm, and a geometric width of 1.4. Ice nucleates in these particles around a T-dependent, nominal threshold RHI of 160%. Here, the nominal freezing threshold is the RHI at which one particle of radius 0.25 µm freezes in one second. Ice nuclei are initialized with 0.3 µm radius and a width of 1.2. The nucleation rates depend on the size-dependent water activity of the freezing particles (shifted to induce immersion freezing at lower RHI in the case of IN) and on T [Kärcher and Lohmann, 2003].

3. Results and Discussion

[10] We obtain statistically robust results by simulating 150 10-day trajectories with different realizations of random wave patterns and sampling data every 30 min for each scenario, HOM or MIX-n_{IN}. We first summarize characteristic features from single 10-day simulations.

[11] In cases HOM (homogeneous freezing only), most clouds form at or after day 4 and have initial ice crystal number concentrations n = 100–4000 L^{-1} and radii r = 1–5 µm. Typically, these clouds deplete [H2O] to saturation. The crystals grow to ~10 µm radius as they fall out of the formation layer, causing RHI to rise again. If the first cloud contains fewer crystals, two or more clouds may form up to day 10. In some cases, ice particles with a total ice water content IWC ≈ 50 µg m^{-2}, equivalent to [H2O] = 0.5 ppmv, can be carried across the tropopause into the lower stratosphere where they would quickly evaporate.

[12] In cases MIX-n_{IN} (IN added to liquid particles), shorter-lived clouds may form very early depending on RH_{IN} and have initial crystal concentrations that are limited by n_{IN}. For n_{IN} = 1 L^{-1}, crystal radii are typically 5–20 µm, and contrary to liquid particles, the IN are completely removed by sedimentation of ice crystals which formed on them. The mean RHI increase driven by the slow cooling is often only slightly retarded by IN-induced SVCs, and such clouds do not reach an equilibrium state [Kärcher, 2002]. The perturbations of the RHI evolution are nevertheless sufficient to change the conditions for homogeneous freezing in the presence of rapid temperature oscillations.

[13] The calculated size distributions are in rough agreement with in situ measurements [Heymsfield, 1986; McFarquhar et al., 2000; Thomas et al., 2002]. The small crystals with r < 5–10 µm mainly form by homogeneous freezing; some of them grow larger when occasionally only a small number of crystals are produced. It is interesting to note that the ice crystal size distribution may extend into the
mean values of $\langle r \rangle$ and $\langle r \rangle$ and hence more effective dehydration. At $n_{IN} = 100 L^{-1}$, $\langle n \rangle$ is about a factor of two smaller than in case HOM, because ice formation is still limited by the number of available IN. This behavior reflects the key indirect effect of IN on cirrus clouds, namely a reduction of $\langle n \rangle$ and a concomitant increase of $\langle r \rangle$. Similar trends have been predicted for midlatitude cirrus [Haag and Kärcher, 2004].

[18] Among all quantities considered, $f_{occ}$ is most susceptible to changes of $n_{IN}$. Figure 2 demonstrates that IN substantially increase cirrus occurrence (by 6% and 16% for RHI$_{IN} = 130\%$ and 110%, respectively) at IN concentrations as low as 0.01 L$^{-1}$. Below 1 L$^{-1}$, $f_{occ}$ increases above the HOM level because lower freezing thresholds are more often surpassed than the high homogeneous thresholds, while at the same time the overall cloud properties in the TTL remain almost unperturbed by the transient clouds formed on IN. Above 10 L$^{-1}$, we enter a regime where cloud properties start to be controlled by IN, as reflected by the steady increase of $\langle r \rangle$, $\langle IWC \rangle$, and $f_{occ}$, and decrease of $\langle [H_2O] \rangle$.

4. Sources of Uncertainties

[16] From Figure 1 we begin to suspect that dehydration and supersaturation might not change drastically by heterogeneous freezing unless a minimum number of efficient IN are present. It is instructive to investigate the mean values (indicated by $\langle \cdot \rangle$) of $[H_2O]$, $r$, and IWC, and the average cirrus cloud occurrence frequency, $f_{occ}$, with the help of Figure 2. The behavior of $\langle S_i \rangle$ follows that of $\langle [H_2O] \rangle$; $\langle S_i \rangle$ takes a value of 1.12 in case HOM (see arrow in Figure 1) and decreases to 1.02 for high $n_{IN}$ and low RHI$_{IN}$. Note that $\langle r \rangle$ and $\langle IWC \rangle$ given below are underestimated, as sedimenting crystals are removed from the trajectories and are therefore underrepresented in the computation of moments of size distributions. This low bias of $\langle r \rangle$ and $\langle IWC \rangle$ does not affect their trends with $n_{IN}$ discussed next. Robust quantitative details about modeled crystal sizes are found at the beginning of this section.

[17] The mean values of $[H_2O]$, $r$, and IWC are not significantly altered compared to scenario HOM unless $n_{IN}$ rises above 0.1–1 L$^{-1}$, regardless of RHI$_{IN}$. In such cases, concerning $\langle [H_2O] \rangle$, it does not matter whether more short-lived clouds with large particles or fewer long-lived clouds with small particles lead to dehydration. For $n_{IN} > 1 L^{-1}$, the mean crystal number density $\langle n \rangle$ (not shown) decreases faster than $\langle IWC \rangle$, leading to larger $\langle r \rangle$ and hence more effective dehydration. At $n_{IN} = 100 L^{-1}$, $\langle n \rangle$ is about a factor of two smaller than in case HOM, because ice formation is still limited by the number of available IN. This behavior reflects the key indirect effect of IN on cirrus clouds, namely a reduction of $\langle n \rangle$ and a concomitant increase of $\langle r \rangle$. Similar trends have been predicted for midlatitude cirrus [Haag and Kärcher, 2004].

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hilation, however, is rather small [Jensen and Pfister, 2004, entry 5 in Table 2].

[21] Ice nuclei may be chemically complex, internally mixed particles. We make no attempt to specify the composition of IN, rather we assume nominal IN freezing thresholds. Values around 130% may not be unrealistic, as the most efficient IN might be removed by wet deposition before entering the TTL. Much lower values might be considered as extreme cases, while values >150% closer to homogeneous freezing thresholds render the IN ineffective in leading to observable changes in cirrus properties. While the properties of individual clouds that form on IN may be depending on the exact IN size distribution and on a possible variability in RHI of IN, our statistical results are still expected to hold in such more complex cases.

[22] We assume that IN are present as separate particles. The effects of IN on cirrus might change upon entrainment into or mixing with other particles, which is difficult to quantify. Further, IN that nucleate ice sediment out of the TTL before reaching the highest altitudes. The effects of IN would increase if they were present throughout the entire TTL.

5. Summary and Conclusion

[23] Our model study suggests that the mean values of ice supersaturation and the stratospheric entry level of H2O are only weakly altered by IN unless they are particularly potent (nominal freezing RHI ≤130%) or abundant (concentrations ≥10 L\(^{-1}\)). Hence, cirrus-induced dehydration may often be controlled by the action of gravity waves and homogeneous freezing of liquid aerosol particles. Additional IN-induced dehydration reaches notable levels of 0.1–0.3 ppmv (up to ~20% of the dehydration potential of cirrus forming by homogeneous freezing) only for IN concentrations 100–1000 L\(^{-1}\). Our current knowledge about IN abundance and properties is insufficient to rule out such an effect.

[24] In contrast, the subvisible cirrus coverage in the TTL is very sensitive to the presence of IN, even at low IN concentrations (0.01–1 L\(^{-1}\)) and modest freezing efficiencies. Heterogeneous IN may more than double the frequency of occurrence of SVCs when present in concentrations 100–1000 L\(^{-1}\), compared to a pure homogeneous freezing scenario. This may have important consequences for the TTL moisture budget and dynamics. Changes in the effective ice crystal radii caused by IN will probably have little effect on the cloud radiative forcing, as the latter is dominated by infrared heating with negligible solar cooling. Changes in ice water content and hence optical depth may be partly compensated by the decreased lifetime of SVCs forming on IN.

[25] The integrated effect of changes in cloud cover and radiative cloud properties in the TTL is difficult to assess and warrants further research, including investigations on characteristics and distributions of small-scale wave-induced temperature perturbations. We support the likely notion that a combination of microphysical (cirrus formation and sedimentation) and dynamical factors (horizontal transport, convection, and waves) control stratospheric humidity and should be treated together in future work.

[26] Neither the microphysical properties of high tropical cirrus nor their impact on radiative fluxes and water vapor are well represented in global models employed to predict climate change. This study emphasizes that more direct IN measurements in the TTL, better emission inventories for potential IN, and a better understanding of IN transport and removal pathways are needed to make headway towards reliable predictions of thin and subvisible cirrus radiative properties and coverage with global models. However, before any cloud changes can be attributed to aerosol effects, a careful evaluation of the dynamical control of cirrus formation is required.

[27] Acknowledgment. I am grateful to George Craig for fruitful discussions and Eric Jensen for providing me with wave-perturbed sample trajectories extracted from meteorological analyses.

References


